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(71) Applicant: **The University of Tokyo**
Bunkyo-Ku, Tokyo (JP)

(72) Inventors:
- **Komiyama, Hiroshi**
  Setagaya-Ku, Tokyo (JP)
- **Osawa, Toshio**
  Shinjuku-Ku, Tokyo (JP)
- **Shirakawa, Hiroaki**
  Kita-Ku, Tokyo (JP)

(74) Representative: **Gibbs, Christopher Stephen**
Haseltine Lake & Co.
Imperial House
15-19 Kingsway
London WC2B 6UD (GB)

(54) A method for forming an ultra microparticle-structure

(57) A method for forming an ultra microparticle-structure composed of ultra microparticles including the steps of:

forming on a substrate (1) higher wettability parts (3) and lower wettability parts (2) to a material to be deposited,

depositing on the substrate (1) the material to be deposited to form particles (4) made of the material on the substrate (1), and

accumulating the particles (4) in the higher wettability parts to form the ultra microparticle-structure composed of the ultra microparticles (5).

FIG. 1a

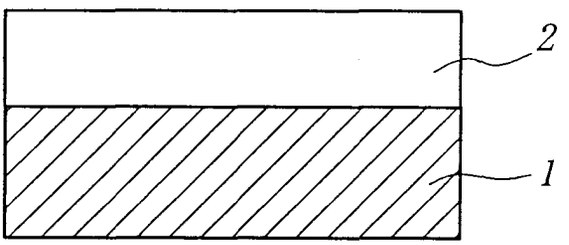

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FIG. 1b

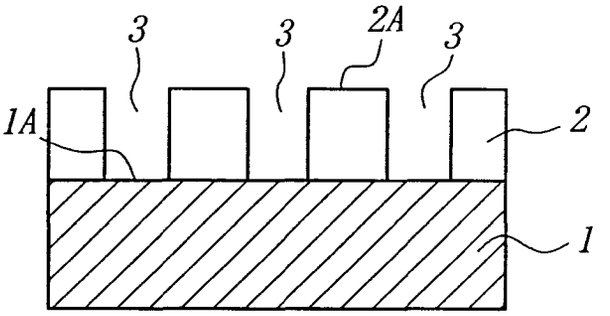


FIG. 1c

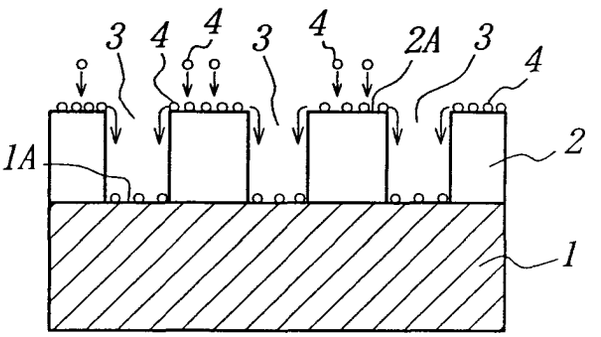
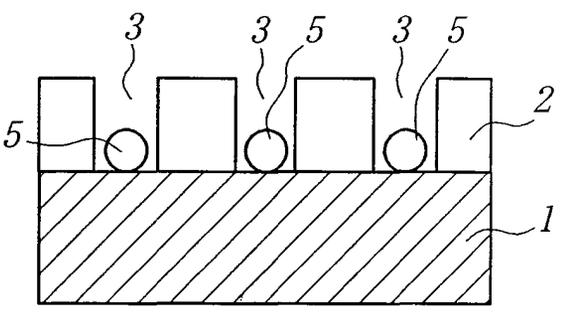


FIG. 1d



Description

[0001] This invention relates to a method for forming an ultra microparticle-structure, and more particularly such a method suitable for forming quantum well wires and quantum well dots in semiconductor micro-processing techniques.

[0002] Recently, semiconductor micro-processing techniques have been rapidly developing and large-scale integrations (LSIs) having feature dimensions of 300 nm have been realized.

[0003] On the other hand, when electrons are confined within a very small area having a dimension of several tens of nm in a semiconductor, the quantum nature of the electron become conspicuous. Thus, taking advantage of this nature, a new functional device such as an electron interference wave element or a single electron element which operates on individual electrons can be realized.

[0004] Moreover, if a low-dimensional quantum structure such as quantum well wires or quantum well dots is applied to an active layer of a semiconductor laser, for example, it is theoretically predicted that the characteristics of the laser are remarkably enhanced, compared with a conventional semiconductor laser.

[0005] Thus, it is strongly desired to realize nm-scale processing techniques in the field of semiconductor micro-processing technology.

[0006] To pursue the above object, in forming semiconductor-quantum well wires or quantum well dots, for example, a method has been developed in which a quantum well structure is formed through crystal growth on a wafer, which is processed in strip and rectangle by lithography and etching, and the side walls of the processed wafer are embedded by secondary crystal growth.

[0007] Similarly, in forming a low-dimensional quantum structure such as semiconductor-quantum well wires or quantum well dots, methods using a crystal-growing technique have been developed, such as: (1) a method in which a strip structure is formed by using a step flow mode to grow a crystal in a transverse direction from an atomic face-step in a slightly slipped surface (slightly inclined face) to the low index face of the crystal, (2) a method in which, after a three-dimensional structure with small faces known as "facets" is selectively formed in openings of a substrate partially covered with amorphous film, on a part of the three-dimensional structure, a quantum well wire- or a quantum well dot-structure, is formed, the upper surface of which is covered with another semiconductor crystalline film, (3) a method in which, in crystal-growing on a substrate of which a surface is processed to have a concave-convex shape, a quantum well wire- or a quantum well dot-structure is formed at a given position on the substrate by using different crystal growth rate and is covered with another semiconductor crystalline film, (4) a method in which quantum well dots are self-formed using distortion between heterojunction with different lattice constant, without specific processing of the crystalline underlayer.

[0008] However, the former method using the crystal processing of lithography restricts a lateral confining size and brings about defects in a re-growing boundary.

[0009] Moreover, the latter methods using crystal growth techniques tend to degrade through a step bunching phenomena or a step ordering phenomena.

[0010] Furthermore, the above methods can miniaturize the quantum well wires or quantum well dots only down to several tens of nm-scale and cannot provide ones with an ideal scale of several nm.

[0011] It is therefore desirable to provide a method for forming an ultra microparticle-structure composed of ultra microparticles of several nm-scale.

[0012] It is also desirable to provide an alternative method for forming low dimensional quantum structures.

[0013] This invention relates to a method for forming an ultra microparticle-structure composed of ultra microparticles comprising the steps of:

forming on a substrate parts respectively having higher wettability and lower wettability with respect to a material to be deposited,
depositing on the substrate the material to be deposited to form particles made of the material on the substrate, and
accumulating the particles in the higher wettability parts to form the ultra microparticle-structure composed of the ultra microparticles.

[0014] Fig. 1 is a general view for explaining one method for forming an ultra microparticle-structure according to the present invention.

[0015] First of all, as shown in Fig. 1(a), for example, a film 2 made of an energically unstable material with respect to a material to be deposited is formed on a substrate 1 to lower the wettability of the whole surface of the substrate 1 with respect to (i.e. as wetted by) the material to be deposited.

[0016] Next, as shown in Fig. 1(b), openings 3 are formed on the surface of the substrate 1 to form thereon higher wettability parts with respect to the material to be deposited.

[0017] Thereafter, a target made of the material to be deposited is sputtered and particles 4 made of the material are deposited on the substrate 1 at a deposition rate of 0.01-10 nm/sec (over the whole substrate).

[0018] At this stage, the surface of the substrate 1 has lower wettability parts for the particles 4 due to the film 2 formed thereon. Thus, as soon as the particles 4 are deposited on the substrate 1 (surfaces 2A of the film 2), they immediately migrate along the surface (the surfaces 2A of the film 2) of the substrate 1. Then, when they come to the openings 3 having higher wettability, they fall into the openings to become more energetically stable. Therefore, the particles 4 accumulate in the open-

ings 3 and, lastly, as shown in Fig. 1(d), they collide and coalesce with one another to form ultra microparticles 5.

[0019] In this case, some particles 4 may melt upon collision. Such melting can make the ultra microparticles 5 substantially spherical.

[0020] The sizes of the ultra microparticles 5 formed as described above are determined within (are limited by) the sizes of the openings 3. Moreover, they can be freely controlled within the sizes of the openings 3 by adjusting the amount of the particles 4 to be deposited and changing the degree of migration of the particles 4.

[0021] Therefore, the ultra microparticle-structure composed of ultra microparticles of several nm-scale is easily formed, this being a desirable result.

[0022] The invention will be more particularly described by way of example, with reference to the accompanying drawings, in which:

Figs. 1(a) to (d) are general views for explaining a method for forming an ultra microparticle-structure according to the present invention,

Figs. 2(a) and (b) are TEM photographs of an example of an ultra microparticle-structure formed by a forming method according to the present invention,

Fig. 3 is a TEM photograph of another example of an ultra microparticle-structure formed by the forming method according to the present invention,

Fig. 4 is a TEM photograph of a further example of an ultra microparticle-structure formed by a forming method according to the present invention, and

Fig. 5 is a TEM photograph showing a growing process of ultra microparticles constituting an ultra microparticle-structure according to the present invention.

[0023] The invention will be described in detail as follows, with reference to the above drawings.

[0024] Figs. 1 (a) to (d) are general views for explaining the forming method of an ultra microparticle-structure according to the present invention, as above-mentioned.

[0025] In the forming method of an ultra microparticle-structure, higher and lower wettability parts to a material to be deposited are needed to be formed on a surface of a substrate 1.

[0026] In the case of forming only the lower wettability parts on the surface of the substrate 1, when particles 4 made of the material to be deposited strike the surface, they do not accumulate thereon, and ultra microparticles 5 are not formed.

[0027] In the case of forming only the higher wettability parts on the surface of the substrate 1, when particles 4 made of the material to be deposited strike the surface, they become energetically stable without sufficient migration. Thus, as is the case with forming only the lower wettability parts, the particles 4 do not accumulate to form the ultra microparticles 5.

[0028] The low wettability parts can be formed on the surface of the substrate 1 by forming a film 2 made of a lower wettability material to the material to be deposited on the surface through evaporation, sputtering, CVD, MBE, MOVPE, etc. Moreover, the parts may be formed through ion platting, laser abrasion. Alternatively, those parts may be formed by making the substrate 1 itself of the lower wettability material.

[0029] The high wettability parts can be formed on the surface of the substrate 1 by forming openings 3 in the film 2 made of a lower wettability material to a material to be deposited or in the substrate, as above-mentioned. In this latter case, the physical characteristic of the opening itself, rather than the higher wettability material revealed by the openings, leads to higher wettability. In addition, the openings may be formed through ion-irradiation, electron beam irradiation or ultraviolet-irradiation of the surface of the substrate. Moreover, they may be formed by applying a solution, radical treatment, plasma treatment, partial heating of infrared laser, or plating.

[0030] The openings 3 may be formed through plasma etching, vapor phase reaction (dry-etching), or liquid phase reaction (wet-etching) to the surface of the film 2 or the substrate 1.

[0031] The size of the openings 3 is determined, depending upon the size of the ultra microparticles to be formed. For forming nm-scaled ultra microparticles easily, it is preferably 0.5-100 nm, particularly 3-30 nm.

[0032] The material to be deposited may be deposited onto the substrate by a sputtering method, an evaporation method, a CVD method, a MBE method, or a MOVPE method. These methods may be selected in accordance with the sort of the material or other reasons.

[0033] The sputtering method may be preferably employed for the deposition because it can easily control its deposition rate. In this case, by setting the deposition rate over 1 nm/sec, the particles 4 made of the material are likely to melt. Consequently, ultra microparticles 5 with substantially spherical shape may be formed.

[0034] In the case of constituting the openings 3 (the higher wettability parts to the material to be produced on the surface of the substrate) as above-mentioned, the size of the ultra microparticles 5 can be controlled within that of the openings 3 by appropriately adjusting the amount of the particles 4 made of the material to be deposited or adjusting the migrating amount of the particles 4 towards the openings 3.

[0035] The amount of particles 4 may be controlled by adjusting a sputtering rate in sputtering the material to be deposited, for example. In the case of employing another deposition method, it may be controlled by changing an evaporation rate or a reaction time of the material.

[0036] Moreover, it may be controlled by interposing a mesh between a source such as a target and the substrate or making a target of amorphous material.

[0037] The migration of the particles 4 to the openings 3 with high wettability may be controlled by changing the

deposition rate of the material to be deposited as above-mentioned or by changing the energies of the particles 4 via the change of the substrate temperature, that is, the kinetic energies of the particles 4 on the substrate.

[0038] Moreover, it may be controlled by adjusting the wettability of the surface of the substrate and adjusting the flatness of the substrate.

[0039] The particles 4 migrate on the surface of the substrate, that is, on a surface 2A of the film 2 in Fig. 2, and collide and coalesce with one another on the surface 2A and a surface 1A of the substrate 1 inside the openings 3 to increase their sizes.

[0040] One can embed the openings 3 on the substrate 1 to prevent the particles 4 from falling into the openings. The covering of the particles 4 prevents the particles from further colliding and coalescing with one another. Consequently, the embedding and the covering prevent the size of the deposited particles 4 from increasing beyond the size desired.

[0041] The material used to embed the openings 3 or cover the particles 4 is not restricted, and any kind of material may be employed for embedding or covering the particles. For forming the ultra microparticle-structure uniting the substrate as shown in Figs. 1 (a) to (d), it is desirable to use the same material as for the film 2 or the substrate 1.

[0042] For example, the above-obtained ultra microparticle-structure has ultra microparticles 5 as shown in Fig. 1(d). Sizes of 0.5 nm to 100 nm, preferably 3 nm to 30 nm, are achievable by controlling the degree of migration.

[0043] Figs. 2 to 4 are TEM photographs showing an ultra microparticle structure formed by the forming method according to the present invention.

[0044] Figs. 2 to 4 each show ultra microparticle-structures in which particles made of Au are deposited onto a substrate made of amorphous SiO_2. In these Figures, each dark point represents an ultra microparticle formed in an opening.

[0045] Figs. 2(a) and (b) show ultra microparticle-structures formed on a substrate kept at room temperature, and Figs. 3 and 4 show ultra microparticle-structures formed on a substrate kept at 500°C.

[0046] Figs. 2(a) and (b) show ultra microparticle-structures formed by depositing the particles for 20 seconds and 60 seconds, respectively. Moreover, Fig. 4 shows an ultra microparticle-structure formed by embedding openings in the same amorphous SiO_2 as the material composing the substrate after depositing the particles as shown in Fig. 3.

[0047] Since these TEM photographs are taken at 400,000 times magnification, from measuring the size of the particles in the photographs, it can be seen that they have a size of about 2-4 nm, which is a desired size. That is, the ultra microparticle-structure formed by the forming method of an ultra microparticle-structure of the present invention has ultra microparticles of nm-scale.

[0048] Thus, since the ultra microparticle-structure has particles with ideal sizes for forming quantum well wires and quantum well dots, it is expected to be suitable for realizing ones of nm-scale.

[0049] This invention is now concretely described in the following examples, with reference to the drawings.

Example 1

[0050] In this example, the substrate 1 itself was made of a material having a lower wettability to the material to be deposited, without forming the film 2 as shown in Fig. 1. The material to be deposited was Au, and the material having the lower wettability was amorphous SiO_2. Higher wettability parts were provided as openings. These had a diameter of 3-30 nm and were formed in the SiO_2 layer by ion beam equipment. The substrate 1 was composed of a support member to give the material sufficient strength, here a Cu-mesh, and a layer made of the amorphous SiO_2 formed on the Cu-mesh by sputtering. Alternatively, sintered amorphous SiO_2 could be used, without requiring an additional support layer.

[0051] The material to be deposited was deposited, at a deposition rate of 0.016 nm/sec by sputtering, on the substrate 1 kept at room temperature.

[0052] When ultra microparticle-structures formed at deposition-times of 20 seconds and 60 seconds were observed by a TEM, respectively, the ultra microparticle-structures turned out to have structures shown in Figs. 2(a) and (b).

[0053] When the average sizes of the ultra microparticles constituting the ultra microparticle-structures were measured from the 400,000 times-TEM photographs shown in Figs. 2(a) and (b), they were 2.10 nm and 3.64 nm with a 20 seconds-deposition time in Fig. 2(a) and a 60 seconds-deposition time in Fig. 2(b), respectively.

Example 2

[0054] Except that the temperature of the substrate was 500°C and the deposition time was 60 seconds, an ultra microparticle-structure was formed as in Example 1. When the thus obtained ultra microparticle-structure was observed by a TEM, it turned out to have a structure as shown in Fig. 3.

[0055] When the average size of the ultra microparticles constituting the ultra microparticle-structure was measured as in Example 1, it was 3.37 nm.

Example 3

[0056] An ultra microparticle-structure was formed as in Example 2 and was held on the substrate 1 kept at 500°C for 2 hours.

[0057] The thus obtained ultra microparticle-structure was observed by a TEM, it turned out to have a structure as shown in Fig. 4.

[0058] When the average size of the ultra microparticles constituting the ultra microparticle-structure was observed by the same manner as in the above examples, it turned out to be 3.64 nm.

Example 4

[0059] In this example, an ultra microparticle-structure was formed as in Example 2, except that Si was used as the support layer of the substrate. After the ultra microparticles constituting the ultra microparticle-structure were covered with the same amorphous SiO_2 material as the substrate 1, the ultra microparticle-structure was observed at a cross section perpendicular to the surface of the substrate 1. Consequently, it turned out to have a structure, at the cross section, as shown in Fig. 5.

[0060] As is apparent from Examples 1 to 3, according to the forming method of an ultra microparticle-structure of the present invention, an ultra microparticle-structure composed of ultra microparticles on a nm-scale can be formed.

[0061] Moreover, when the deposition time of the material to be deposited is increased in Example 1 or the substrate temperature is increased in Example 2, the sizes of the ultra microparticles constituting the ultra microparticle-structure are increased.

[0062] From the fact that the ultra microparticles are uniformly dispersed in Example 2, on the contrary, they are randomly dispersed in Example 3, it is apparent that they migrate during the two hours. Moreover, as is apparent from the TEM photograph in Example 4, the ultra microparticles, constituting the ultra microparticle-structure formed by the forming method of the present invention, are almost spherical.

[0063] As above-mentioned, amorphous SiO_2 material is employed as the lower wettability material and Au material is employed as the material to be deposited in the examples. Then, the substrate comprises a layer made of the amorphous SiO_2. However, the present invention may be applied not only with these materials, but also for any kind of material.

[0064] For example, as the material to be deposited may be used Si, GaAs, InP, GaN, ZnS, ZnO, AlN, Al2O3, TiO_2, $PbTiO_3$, W, Ag, Cu, $CuCl_2$, CdS, PbS, a compound of these materials, etc., in addition to the above Au.

[0065] Moreover, as the lower wettability material constituting the substrate may be used Si, amorphous Si, amorphous SiH_x, $Si3N_4$, amorphous SiN, $Al2O_3$, CaF_2, NaCl, TiN, SiC, GaAs, AlN, Zn, TiO_2, graphite, diamond, etc., besides the above amorphous SiO_2.

[0066] Furthermore, in forming the film 2 as shown in Fig. 1, if required, a lower wettability material to the material to be deposited is selected from among the above materials, and the film 2 is formed of the selected material by sputtering.

[0067] Although the present invention has been described in detail with reference to the above examples, this invention is not limited to the above disclosure and every kind of variation and modification within the claim scope may be made without departing from the scope of the present invention.

[0068] As mentioned above, according to the present invention, an ultra microparticle-structure composed of ultra microparticles with several nm-scale can be easily formed. As a result, quantum well wires or quantum well dots having a low-dimensional quantum structure may be formed in an ideal size. Moreover, a device having new functions such as a single-electron element can be also formed.

[0069] Furthermore, VLSIs can be much miniaturised by using the present invention.

Claims

1. A method for forming an ultra microparticle-structure composed of ultra microparticles comprising the steps of:

   forming on a substrate (1) higher wettability parts (3) and lower wettability parts (2) with respect to a material to be deposited,
   depositing on the substrate the material to be deposited to form particles (4) made of the material on the substrate, and
   accumulating the particles in the higher wettability parts (3) to form the ultra microparticle-structure composed of the ultra microparticles (5).

2. A method for forming an ultra microparticle-structure as defined in claim 1, wherein the higher wettability parts (3) are composed of openings formed on a surface of the substrate.

3. A method for forming an ultra microparticle-structure as defined in claim 1 or 2, wherein the material deposited is deposited by sputtering.

4. A method for forming an ultra microparticle-structure as defined in claim 3, wherein the deposition rate of the material deposited is 0.01 nm/sec to 10 nm/sec.

5. A method for forming an ultra microparticle-structure as defined in any one of claims 1 to 4, wherein the sizes of the ultra microparticles (5) constituting the ultra microparticle-structure are controlled by adjusting the amount of the material deposited.

6. A method for forming an ultra microparticle-structure as defined in claim 5, wherein the amount of the material deposited is controlled by adjusting the deposition rate of the material.

7. A method for forming an ultra microparticle-structure as defined in any one of claims 1 to 6, wherein the sizes of the ultra microparticles (5) constituting the ultra microparticle-structure are controlled by adjusting the temperature of the substrate.

8. A method for forming an ultra microparticle-structure as defined in any one of claims 1 to 7, wherein the ultra microparticles (5) constituting the ultra microparticle-structure are formed by collision and coalescence of the particles deposited by migration of the particles (4) on the substrate.

9. A method for forming an ultra microparticle-structure as defined in claim 2, further comprising the steps of:

embedding the openings (3) formed on the surface of the substrate after depositing the material to be deposited on the substrate, and covering the ultra microparticles (5),
whereby the size of the ultra microparticles (5) constituting the ultra microparticle-structure is prevented from being increased.

10. A method for forming an ultra microparticle-structure as defined in any one of claims 1 to 9, wherein the wettability of the surface of the substrate (1) is lowered by directly covering the surface at least partially with a material of relatively low wettability with respect to the material to be deposited.

11. An ultra microparticle-structure composed of approximately spherical ultra microparticles (5) having diameters of 0.5 nm to 100 nm.

12. A low-dimensional quantum structure, such as quantum well wires or dots, incorporating an ultra microparticle-structure as defined in claim 11.

FIG. 1a

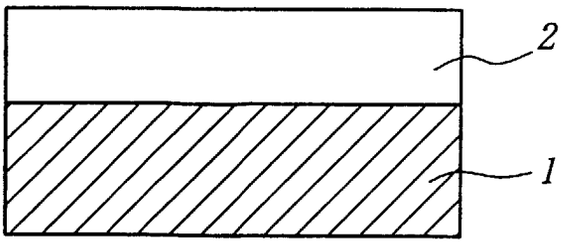


FIG. 1b

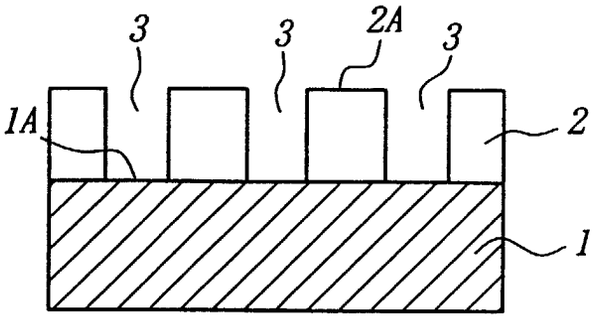


FIG. 1c

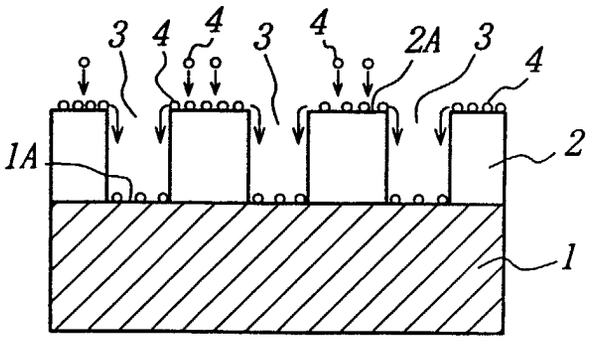
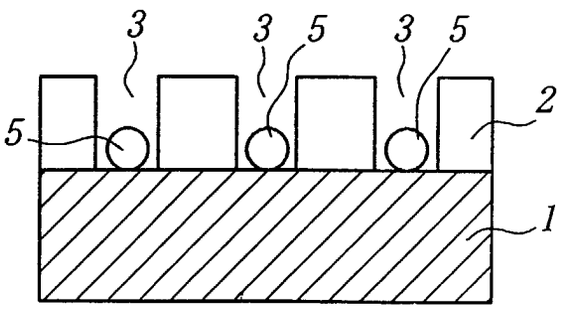


FIG. 1d



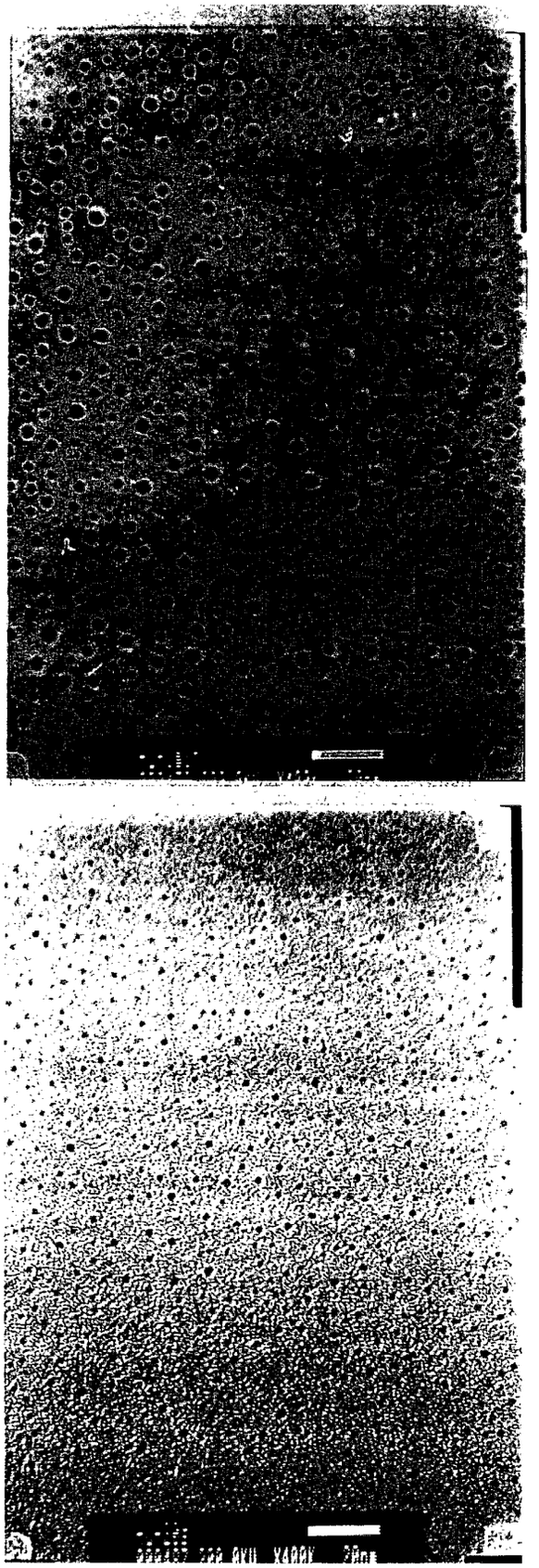


FIG. 2

FIG. 3

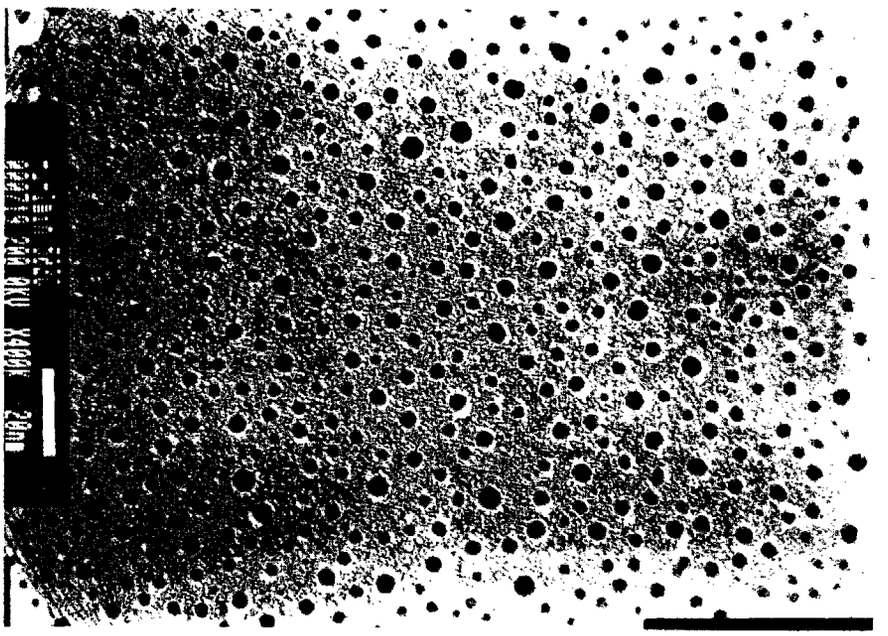


FIG. 4

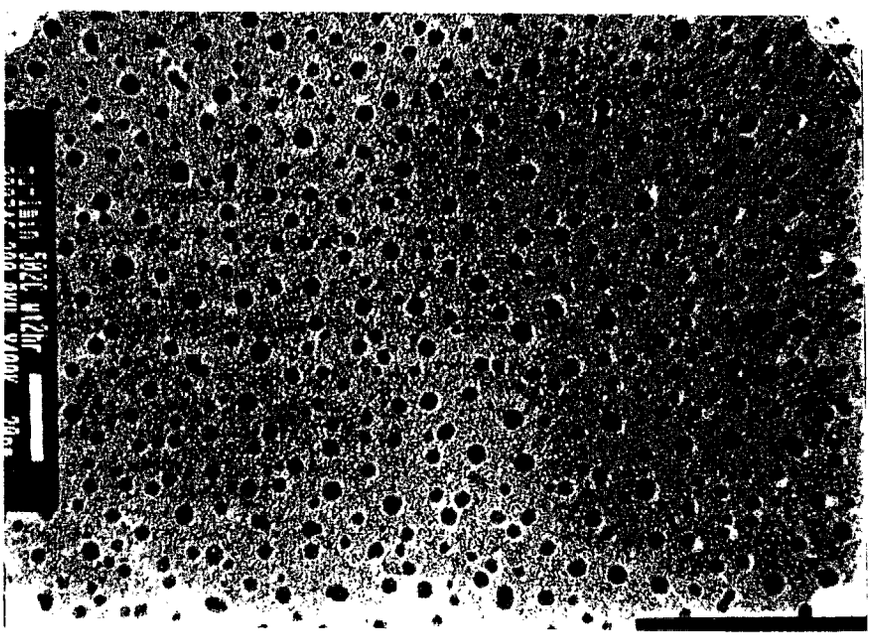
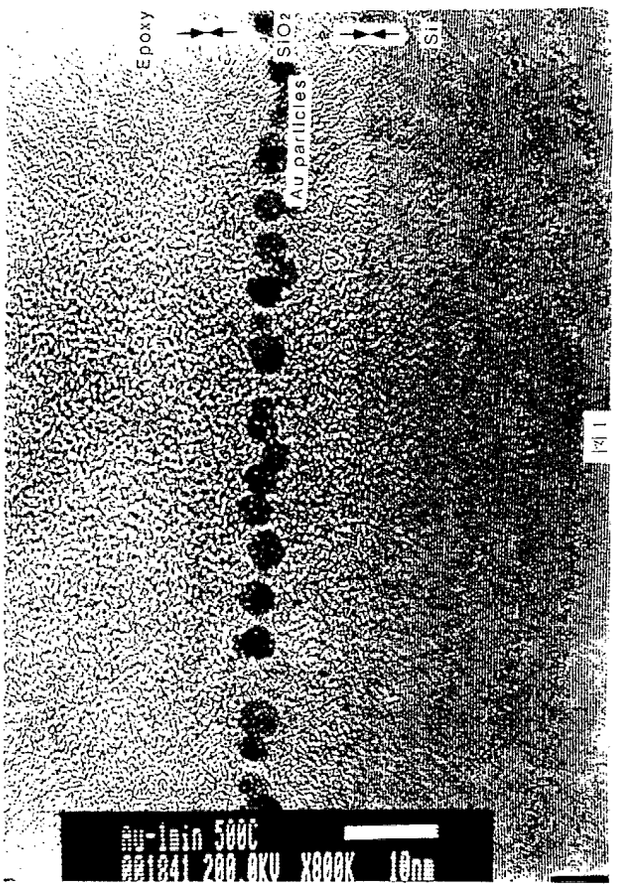


FIG. 5


Epoxy
SiO2
Au particles
Au particles
Si
au-1min 500C
001041 200.0KV X800K 10nm

European Patent Office

EUROPEAN SEARCH REPORT

Application Number
EP 99 30 8433

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
X	HIRASAWA M ET AL: "Growth mechanism of nanoparticles prepared by radio frequency sputtering" JOURNAL OF APPLIED PHYSICS, 1 AUG. 1997, AIP, USA, vol. 82, no. 3, pages 1404-1407, XP002126273 ISSN: 0021-8979 * figures 1,2 * * paragraph [EXPERIMENT] * * paragraph [RESULTS...] *	1-12	B82B3/00 B82B1/00 H01L29/12 C23C16/04
A	---	1-10	
P,X	SHIRAKAWA H ET AL: "Migration-coalescence of nanoparticles during deposition of Au, Ag, Cu, and GaAs on amorphous SiO/sub 2/" JOURNAL OF NANOPARTICLE RESEARCH, 1999, KLUWER ACADEMIC PUBLISHERS, NETHERLANDS, vol. 1, no. 1, pages 17-30, XP000865746 ISSN: 1388-0764 * figures 1-5,7-9,13 * * tables 1,2 * * paragraph [EXPERIMENTAL...] * * paragraph [RESULTS] *	1-12	
	---		TECHNICAL FIELDS SEARCHED (Int.Cl.7)
X	KOFMAN R ET AL: "Self organized growth and ultrafast electron dynamics of metallic nanoparticles" THIN SOLID FILMS,CH,ELSEVIER-SEQUOIA S.A. LAUSANNE, vol. 318, no. 1-2, page 73-75 XP004138567 ISSN: 0040-6090 * paragraph [0002] *	11,12	B82B H01L C23C H01S
A	---	1-10	
	-/--		

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
BERLIN	17 January 2000	Polesello, P

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

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DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
X	NASU H ET AL: "Preparation and optical properties of semiconductor microcrystal-doped SiO2 glass thin films by rf-sputtering" JOURNAL OF NON-CRYSTALLINE SOLIDS,NL,NORTH-HOLLAND PHYSICS PUBLISHING. AMSTERDAM, vol. 178, page 148-154 XP004067766 ISSN: 0022-3093 * figures 3-6 * * paragraphs [0002],[0003] *	11,12	
A	---	1-10	
X	STELLA A ET AL: "Raman versus interband absorption in Ge quantum dots" THIN SOLID FILMS,CH,ELSEVIER-SEQUOIA S.A. LAUSANNE, vol. 318, no. 1-2, page 100-102 XP004138573 ISSN: 0040-6090 * table 1 * * page 100, column 1, line 17 - column 2, line 21 *	11,12	
A	---	1-10	TECHNICAL FIELDS SEARCHED (Int.Cl.7)
X	HIRASAWA M ET AL: "Synthesis of GaAs nanoparticles by digital radio frequency sputtering" APPLIED PHYSICS LETTERS, 4 DEC. 1995, USA, vol. 67, no. 23, pages 3483-3485, XP002126275 ISSN: 0003-6951 * figures 1,2 * * page 3483, column 1, line 34 - column 2, line 32 *	11,12	
A	-----	1-10	

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
BERLIN	17 January 2000	Polesello, P

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

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